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Acknowledgments

C.H. Mueller acknowledges the National Research Council fellowship program. The authors thank N. Varaljay, B. Viergutz, and L. McQuaid for the patterning and fabrication of phase shifters.

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A COMPARISON OF MOCLD WITH PLD $\text{Ba}_x\text{Sr}_{1-x}\text{TiO}_3$ THIN FILMS ON LaAlO_3 FOR TUNABLE MICROWAVE APPLICATIONS

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Historically, tunable dielectric devices using thin crystalline $\text{Ba}_x\text{Sr}_{1-x}\text{TiO}_3$ (BST) films deposited on lattice-matched substrates, such as LaAlO_3 , have generally been grown using pulsed laser deposition (PLD). Highly oriented BST films can be grown by PLD but large projects are hampered by constraints of deposition area, deposition time and expense. The Metal-Organic Chemical Liquid Deposition (MOCLD) process allows for larger areas, faster turnover and lower cost. Several BST films deposited on LaAlO_3 by MOCLD have been tested in 16 GHz coupled microstrip phase shifters. They can be compared with many PLD BST films tested in the same circuit design. The MOCLD phase shifter performance of 293° phase shift with $53 \text{ V}/\mu\text{m}$ dc bias and a figure of merit of $47^\circ/\text{dB}$ is comparable to the most highly oriented PLD BST films. The PLD BST films used here have measured XRD full-width-at-half-maxima (FWHM) as low as 0.047° . The best FWHM of these MOCLD BST films has been measured to be 0.058° .

INTRODUCTION

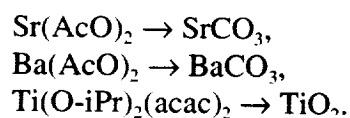
Voltage tunable high-frequency devices using thin $\text{Ba}_x\text{Sr}_{1-x}\text{TiO}_3$ (BST) films have been intensively investigated during the last decade. One approach involves using very thin films of BST, generally of thickness $<50 \text{ nm}$, in parallel plate capacitors. This approach has the advantages of small size, low voltages and easy integration but faces obstacles from losses due to interface and electrode effects. Typically MOCVD BST films are used for parallel plate capacitors [1,2]. A second approach uses larger structures where voltage is applied between electrodes parallel to the surface of the BST film. Examples of this 'planar electrode' approach include gap capacitors, interdigital capacitors, coplanar waveguide and coupled microstrips. An advantage of this configuration is that the BST can be grown on a lattice-matched crystalline substrate, allowing highly oriented BST crystalline thin films to be grown. The BST films used in this configuration must be thicker, usually $>300 \text{ nm}$, in order to tune effectively. The second approach to date has achieved higher microwave figures of merit but at the cost of higher voltages. Compare, for instance, the best reported phase shift per dB loss for a parallel plate integrated phase shifter design which has just recently increased to $60^\circ/\text{dB}$ [2] from a previous

high of 27.1°/dB [3], with the best planar varactor-based phase shifters which have shown up to 110°/dB [4].

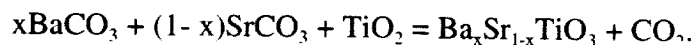
The planar electrode tunable high frequency devices have typically used BST films deposited by pulsed laser deposition (PLD). While high quality crystalline BST films can be routinely achieved by PLD, this deposition method has obvious drawbacks for building large numbers of devices for manufacturing or even demonstrating manufacturability using many device prototypes. PLD tends to be time consuming, expensive (involving large vacuum systems), usually has small deposition areas, and lacks easy repeatability between deposition runs. Other thin film BST deposition methods have been tested in this configuration, including sputtering, chemical combustion vapor deposition (CCVD) and recently sol-gel. In this poster, we describe measurements done on Metal-Organic Chemical Liquid Deposition films (MOCLD) which show similar phase shifter performance as PLD films and adequate crystalline quality but have greater promise for manufacturing.

EXPERIMENTAL DETAILS:

The BST films discussed here were deposited either by pulse laser ablation at a number of different sources including universities, NASA and industry, or were MOCLD films produced by Corning Applied Technology (formerly NZ Applied Technologies). All of the BST films were deposited on 254 μm thick (100) single crystal LaAlO_3 substrates. The majority of these films had Ba:Sr ratios of either 50:50 but or 60:40 and two had a ratio of 40:60. The higher Ba content samples have higher Curie temperatures usually leading to higher tuning and loss at room temperature. The MOCLD $\text{Ba}_{0.5}\text{Sr}_{0.5}\text{TiO}_3$ films were deposited on two LaAlO_3 wafers with a BST thickness of 500 nm. In the MOCLD process, barium acetate ($\text{Ba}(\text{AcO})_2$), strontium acetate ($\text{Sr}(\text{AcO})_2$), and titanium diisopropyl bisacetylacetonate ($\text{Ti}(\text{O-iPr})_2(\text{acac})_2$) were used as precursors. These compounds were dissolved in a water-ethanol mixture. After dipping a substrate into the solution and subsequently withdrawing it, the resulting film was calcined to remove the solvent and to decompose the organic compounds:



This oxide precursor film was then annealed at 650 to 800 °C to form a BST film,



The $\text{Ba}_x\text{Sr}_{1-x}\text{TiO}_3$ films grown on LaAlO_3 substrates using the MOCLD technique were c-axis oriented. Atomic force microscopic (AFM) analysis indicated that the surface roughness of the film was around 20 Å. The surface roughness (as well as the grain size) of the films was observed to increase when the growth temperature increased.

After BST deposition, the films were metallized using electron-beam evaporation at NASA Glenn Research Center with a 15 nm chrome (Cr) or titanium (Ti) adhesion layer followed by a 1.7 to 2.2 μm thick Au film. Standard lift-off chemical etching techniques were used to

fabricate the Au/BST/substrate phase shifters. Finally, a (Cr or Ti)/Au ground plane with the same thicknesses was e-beam evaporated on the back of the substrate.

The phase shifter design consists of n -coupled microstrip sections in series. The phase shift is due to tuning the dielectric constant of the BST in the gap (ϵ_{rBST}) between microstrips. The relation between phase shift and ϵ_{rBST} is nonlinear, with differential phase shift increasing at lower operating frequencies. Each section functions as a single pole broadband filter whose passband shifts with dc bias applied to the ferroelectric. The phase shift is proportional to n . Measurements were done on one coupled microstrip phase shifter design; an eight-coupled section design on 254 μm thick LaAlO_3 with a total length of 1 cm. A cross-sectional view of a phase shifter is shown in Fig. 1 and a schematic representation is shown in Fig. 2. The dimensions are $l = 470 \mu\text{m}$, $s = 7.5 \mu\text{m}$, and $w = 25 \mu\text{m}$. These phase shifters are fairly narrowband, about 12% bandwidth, and the optimal frequency of operation, f_{opt} , depends upon

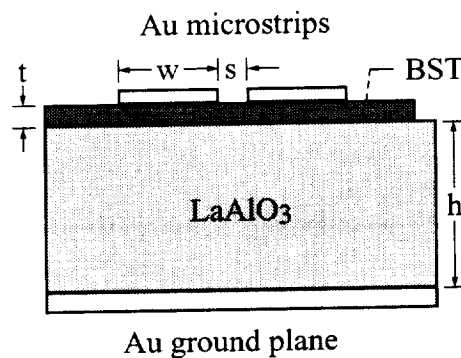


FIGURE 1 Cross-section of a couple microstrip phase shifter section. $w = 25 \mu\text{m}$, $s = 7.5 \mu\text{m}$, $h = 254 \mu\text{m}$, $t = 0.3$ to $1.4 \mu\text{m}$.

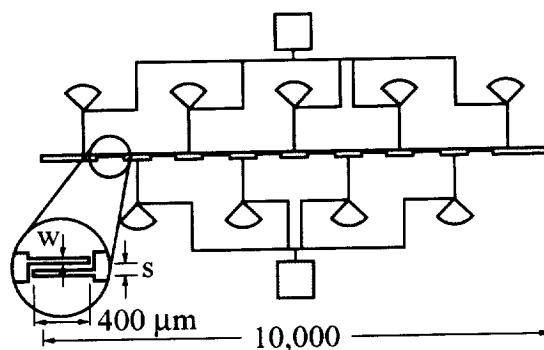


FIGURE 2 Schematic of eight element coupled microstrip phase shifter on LaAlO_3 . $w = 25 \mu\text{m}$, $s = 7.5 \mu\text{m}$.

the ϵ_r and thickness of the ferroelectric film. A detailed discussion of the device properties has been given elsewhere [5]. One should note that since all phase shifters in this study were identical, the BST films may be compared irrespective of the phase shifter design.

RESULTS

Figures 3 to 5 show the performance of the three phase shifters on MOCLD BST that were tested. Figures 3 and 4 show phase shifters fabricated using the first BST coated LaAlO_3 wafer, while Fig. 5 shows the phase shifter fabricated on the second wafer. The figures show both loss phase shift and versus applied dc voltage. Data were taken at 16 GHz. The phase shift observed is near the maximum measured for this design of phase shifter. The variation between phase shifters is small, $<5\%$ on the phase shift at 400 V bias. The variation in loss is also acceptably small. The figures of merit of phase shift per dB loss (K-factor) are 47, 44 and $45^\circ/\text{dB}$, respectively.

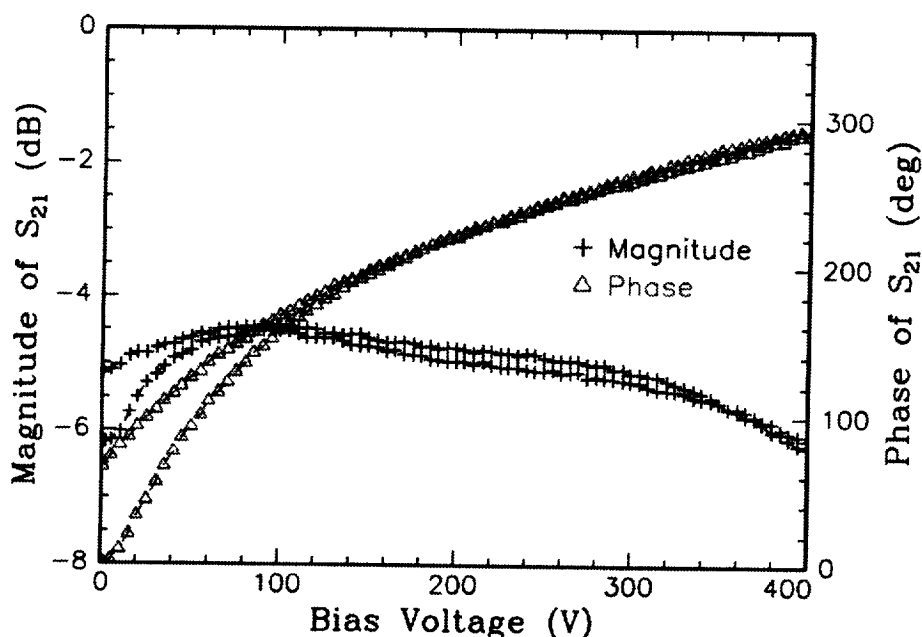


FIGURE 3 Magnitude and Phase Shift of S_{21} vs. applied dc bias of the first phase shifter patterned on 500 nm thick MOCLD $\text{Ba}_{0.5}\text{Sr}_{0.5}\text{TiO}_3$, BST film #1. $T = 300\text{ K}$, $f = 16\text{ GHz}$.

8 element CMPS Au/Ti/BSTO/.01" LAO
 Film: NZAT #1D MOCLD 500 μm BST 50:50, T = 300 K, f=16 GHz

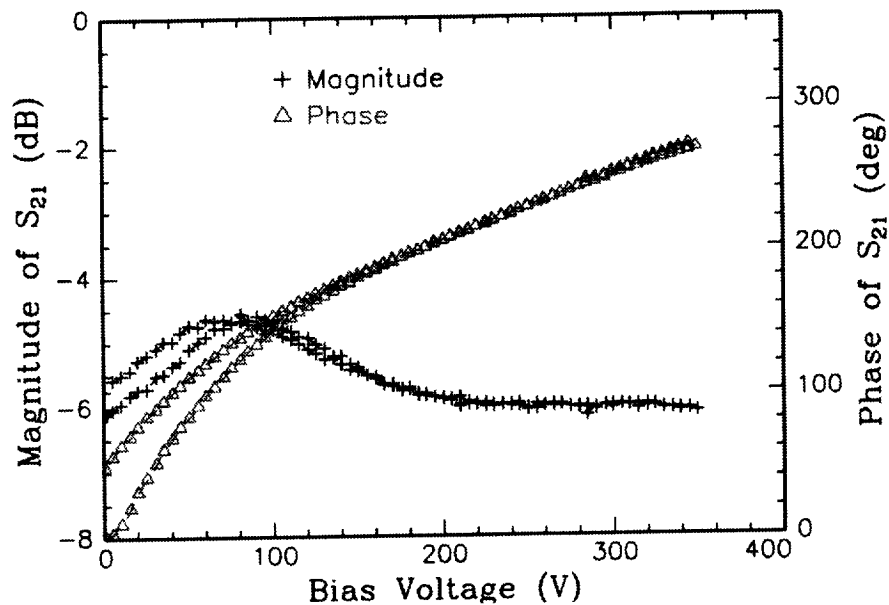


FIGURE 4 Magnitude and Phase Shift of S_{21} vs. applied dc bias of the second phase shifter patterned on 500 nm thick MOCLD $\text{Ba}_{0.5}\text{Sr}_{0.5}\text{TiO}_3$, BST film #1. T = 300 K, f = 16 GHz.

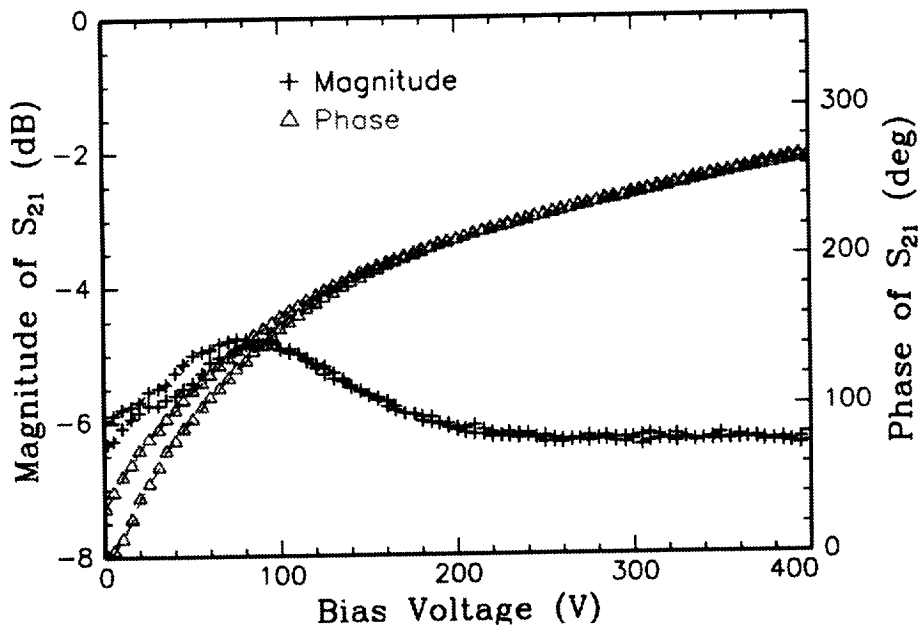


FIGURE 5 Magnitude and Phase Shift of S_{21} vs. applied dc bias of the third phase shifter patterned on 500 nm thick MOCLD $\text{Ba}_{0.5}\text{Sr}_{0.5}\text{TiO}_3$, BST film #2. T = 300 K, f = 16 GHz.

Figure 6 shows the figure of merit of phase shift per dB loss (K-factor) for a large number of phase shifters of the same design. Since the majority of the phase shifter loss is dielectric loss in the BST film, the K-factor roughly corresponds to the ϵ_r tunability per loss tangent at high frequency. The first five film sources all produced BST films using PLD. The first three sources all produced films of thickness 400 nm. Source four's films varied in thickness from 300 to 1400 nm. Source five films were 750 nm in thickness. Sources 1-4 used Ba:Sr ratios of 50:50 or 60:40. Source five produced films with Ba:Sr ratio of 40:60. Source 6 is the MOCLD BST films from Corning Applied Technology Corporation. The MOCLD films are 500 nm thick and have a Ba:Sr ratio of 50:50. The three phase shifters tested on MOCLD films show good repeatability with K-factor and tuning near the high end for all devices tested.

Table I shows some of the other parameters of phase shifter performance along with XRD measurements on some of the samples. The first three columns give the BST film source, Ba:Sr ratio and film thickness. The columns four through eight display the XRD data. The first two red columns are measured a-axis and c-axis lattice parameters in Angstroms. All of the films show higher than ideal bulk lattice parameters. The "percent relaxation" is essentially a measure of the ratio of the c_0 (out-of-plane) to a_0 (in-plane) lattice parameters of the BSTO films. It was calculated using the relation[6]

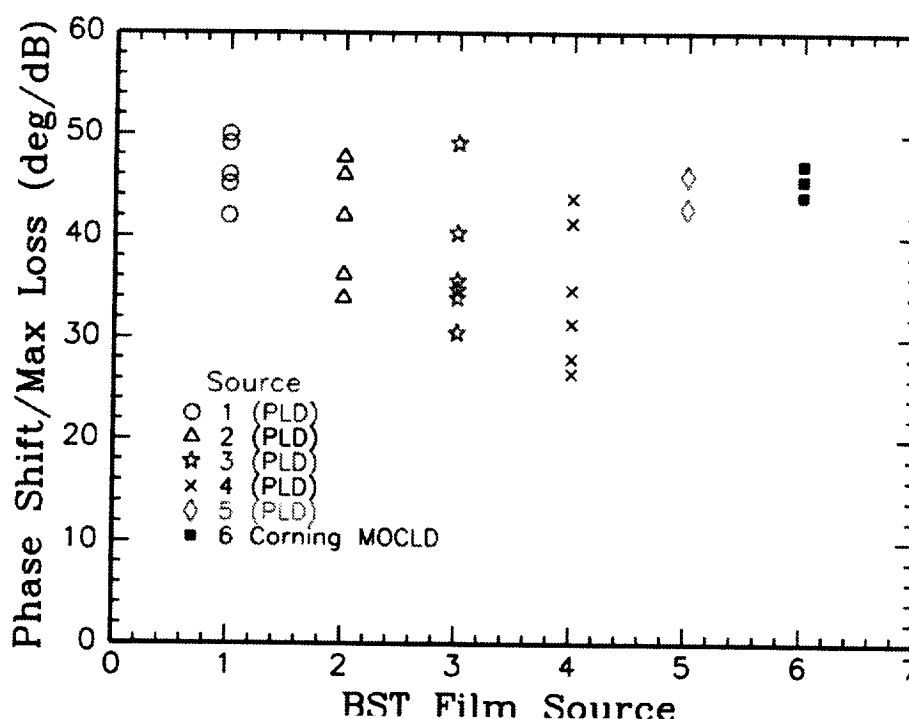


FIGURE 6 Phase Shifter figure of merit of phase shift per dB loss, the K-factor, for identical coupled microstrip phase shifters fabricated on BST films on LaAlO_3 . Films are from six different sources, the first five sources are PLD, source #6 is MOCLD.

TABLE 1 Phase shifter and XRD measurements for many identical coupled microstrip phase shifters fabricated on BST films on LaAlO₃. Films are from six different sources, the first five sources are PLD, source #6 is MOCLD.

Source	Ba/Sr	Thick. nm	a-axis LP	c-axis LP	Percent relaxation	FWHM	Intensity	fopt. GHz	Max phase shift	Applied dc V	Max K. deg/dB
1 (PLD)	50/50	400	3.972	3.966	101.82	368	4218	15	179	150	42
1 (PLD)	50/50	400						15	185	270	46
1 (PLD)	50/50	400						15	179	260	50
1 (PLD)	50/50	400						16	189	400	45.1
1 (PLD)	50/50	400						16	167	400	49.1
2 (PLD)	60/40	400	3.988	3.974	103.5	852	2918	15.5	149	400	46
2 (PLD)	60/40	400	3.977	3.967	102.93	1756	1340	16.5	110.7	350	33.9
2 (PLD)	60/40	400	3.982	3.968	103.86	1848	1532	15.8	130.5	350	36.2
2 (PLD)	60/40	400	3.972	3.958	104.27	790	583	15.8	180	400	47.7
2 (PLD)	60/40	400	3.969	3.948	106.33	930	1728	15.8	195	400	42
3 (PLD)	60/40	400	3.975	3.964	103	306	4022	15.5	281	350	35.6
3 (PLD)	60/40	400	3.976	3.965	103.26	196	8185	15.4	305	400	49.1
3 (PLD)	60/40	400						15.5	298	400	40.3
3 (PLD)	60/40	400	3.973	3.965	102.44	270	4618	15.5	280	350	33.9
3 (PLD)	60/40	400						14	250	200	30.5
3 (PLD)	60/40	400						14.5	305	350	34.7
4 (PLD)	50/50	300	3.961	3.955	101.6	355	4970	14.3	201	400	43.7
4 (PLD)	60/40	1200		3.963	100	900		15	84.4	360	26.5
4 (PLD)	50/50	700	3.965	3.95	104.54	346	8330	15	223	360	34.7
4 (PLD)	50/50	1400	3.974	3.963	100.46	1231	1130	13	192	340	28
4 (PLD)	50/50	800						13.8	130	400	44
4 (PLD)	50/50	500						14.5	135	400	44
5 (PLD)	40/60	750						14	299	400	44
5 (PLD)	40/60	750						13.5	268	324	44
MOCLI	50/50	500						16.3	293	400	44
MOCLI	50/50	500	3.956	3.945	103.39	564	2018	16	273	350	44
MOCLI	50/50	500	3.976	3.952	107	568	2018	16.2	279	400	44

$$\% \text{ relaxation} = \frac{c_{0, \text{measured}} - a_{0, \text{substrate}}}{c_{0, \text{unstrained}} - a_{0, \text{substrate}}} \times 100$$

where a_0 is the lattice parameter of the substrate (for LaAlO₃, assumed to be cubic with a lattice parameter of 3.79 Å), and c_0 is the calculated lattice parameter, assuming a strain-free film. The value for $c_{0, \text{unstrained}}$ is derived from

$$c_{0, \text{unstrained}} = c_{0, \text{measured}} \left[1 + \frac{2\nu}{1+\nu} \right] \left[\frac{c_{0, \text{measured}} - a_{0, \text{measured}}}{c_{0, \text{measured}}} \right]$$

where $a_{0, \text{measured}}$ is the in-plane lattice parameter of the film, and ν is Poisson's ratio (assumed = 0.33). For cubic films in which the interfacial strain is relieved by mechanisms such

as misfit dislocations, $c_0 = a_0$, and thus the %relaxation is 100%. For the BST films studied in this set of experiments, the % relaxation is slightly greater than 100%, which arises because the in-plane lattice parameters of the films are larger than the out-of-plane lattice parameters, despite the fact that the substrate lattice parameter is smaller than that of the films. Thus we conclude that the lattice parameter of the substrate is not the dominant factor which determines film strain; if the substrate were to have a significant impact on film strain, we would expect a_0 to be smaller than c_0 , for the film to be compressively strained, and thus the % relaxation to be less than 100%. The full wave half-maximum (FWHM) in arc-sec and the peak intensity in the next two columns give a good measure of the film crystalline quality. Smaller FWHM correlates well with tuning or phase shift but not necessarily with K-factor (which balances tuning and loss). The percent relaxation for one MOCLD film falls in the middle of normal range for PLD BST films of 101 to 107%, the arc-sec film was measured at the upper end of 107%. The FWHM of the two MOCLD films used for phase shifters was 564 arc-sec, roughly double that of the best PLD, but better than the low-tuning PLD films which ranged up to 1848 arc-sec. However, similar MOCLD BST on LaAlO_3 films from Corning have been measured as low as 210 arc-sec. Lastly, Table I lists phase shifter parameter that were measured for these devices. The first column beyond the XRD measurements lists f_{opt} , optimal frequency of operation for a given phase shifter. Lower values of f_{opt} indicate larger values of ϵ_r and/or film thickness. The maximum phase shift at f_{opt} is next, followed by the dc voltage applied to obtain this phase shift. Lastly, the maximum value of the K-factor is given.

CONCLUSIONS

BST films on LaAlO_3 produced by the MOCLD method nearly equal the crystalline quality of those produced by PLD. Phase shifter performance in the Ku-band (12 to 18 GHz) was found to be as good as high-quality BST films produced by PLD. The MOCLD process has several advantages such as the capacity to fabricate thick films, good control of the stoichiometry, multilayer growth and low processing temperature. This process also makes efficient use of raw materials, is capable of large-area production (up to 6 in.) with no vacuum needed and at relatively low cost. For these reasons, MOCLD shows great promise of manufacturability of BST films for high frequency applications.

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REPORT DOCUMENTATION PAGE			Form Approved OMB No. 0704-0188	
Public reporting burden for this collection of information is estimated to average 1 hour per response, including the time for reviewing instructions, searching existing data sources, gathering and maintaining the data needed, and completing and reviewing the collection of information. Send comments regarding this burden estimate or any other aspect of this collection of information, including suggestions for reducing this burden, to Washington Headquarters Services, Directorate for Information Operations and Reports, 1215 Jefferson Davis Highway, Suite 1204, Arlington, VA 22202-4302, and to the Office of Management and Budget, Paperwork Reduction Project (0704-0188), Washington, DC 20503.				
1. AGENCY USE ONLY (Leave blank)		2. REPORT DATE April 2002		3. REPORT TYPE AND DATES COVERED Technical Memorandum
4. TITLE AND SUBTITLE A Comparison of MOCLD With PLD $\text{Ba}_x\text{Sr}_{1-x}\text{TiO}_3$ Thin Films on LaAlO_3 for Tunable Microwave Applications			5. FUNDING NUMBERS WU-755-08-0B-00	
6. AUTHOR(S) F.W. Van Keuls, C.H. Mueller, R.R. Romanofsky, J.D. Warner, F.A. Miranda, and H. Jiang				
7. PERFORMING ORGANIZATION NAME(S) AND ADDRESS(ES) National Aeronautics and Space Administration John H. Glenn Research Center at Lewis Field Cleveland, Ohio 44135-3191			8. PERFORMING ORGANIZATION REPORT NUMBER E-12783	
9. SPONSORING/MONITORING AGENCY NAME(S) AND ADDRESS(ES) National Aeronautics and Space Administration Washington, DC 20546-0001			10. SPONSORING/MONITORING AGENCY REPORT NUMBER NASA TM-2002-210906	
11. SUPPLEMENTARY NOTES F.W. Van Keuls, C.H. Mueller, R.R. Romanofsky, J.D. Warner, and F.A. Miranda, NASA Glenn Research Center; H. Jiang, Coming Applied Technology Corporation, 14A Gill Street, Woburn, Massachusetts 01801. Responsible person, F.W. Van Keuls, organization code 5620, 216-433-3379.				
12a. DISTRIBUTION/AVAILABILITY STATEMENT Unclassified - Unlimited Subject Categories: 32 and 33 Available electronically at http://gltrs.grc.nasa.gov/GLTRS This publication is available from the NASA Center for AeroSpace Information, 301-621-0390.			12b. DISTRIBUTION CODE	
13. ABSTRACT (Maximum 200 words) Historically, tunable dielectric devices using thin crystalline $\text{Ba}_x\text{Sr}_{1-x}\text{TiO}_3$ (BST) films deposited on lattice-matched substrates, such as LaAlO_3 , have generally been grown using pulsed laser deposition (PLD). Highly oriented BST films can be grown by PLD but large projects are hampered by constraints of deposition area, deposition time and expense. The Metal-Organic Chemical Liquid Deposition (MOCLD) process allows for larger areas, faster turnover and lower cost. Several BST films deposited on LaAlO_3 by MOCLD have been tested in 16 GHz coupled microstrip phase shifters. They can be compared with many PLD BST films tested in the same circuit design. The MOCLD phase shifter performance of 293° phase shift with 53 V/μm dc bias and a figure of merit of 47°/dB is comparable to the most highly oriented PLD BST films. The PLD BST films used here have measured XRD full-width-at-half-maxima (FWHM) as low as 0.047°. The best FWHM of these MOCLD BST films has been measured to be 0.058°.				
14. SUBJECT TERMS Ferroelectric; Tunable; Microwave			15. NUMBER OF PAGES 14	
			16. PRICE CODE	
17. SECURITY CLASSIFICATION OF REPORT Unclassified	18. SECURITY CLASSIFICATION OF THIS PAGE Unclassified	19. SECURITY CLASSIFICATION OF ABSTRACT Unclassified	20. LIMITATION OF ABSTRACT	